Surface Structure of Glasses: Freezing of Capillary Waves?

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Introduction: Thermally excited capillary waves cause lateral correlations from macroscopic down to microscopic length scales on a liquid surface [1,2]. The surface roughness induced by capillary waves typically is of the order of a few ångstroms. We have studied the "freezing" of the capillary wave-induced roughness and the lateral correlations at the glass transition [3,4]. The experimental results are compared with predictions derived from continuum mechanics [5].

Methods and Materials: The surface structure of the prototype glass glycerol has been investigated by surface sensitive x-ray scattering. Macroscopic glycerol films (material purchased from Fluka # 49770, purity better than 99.5%) of approximately 4.5 mm thickness were prepared in a trough of 140 mm diameter. After filling the trough with glycerol, the inner chamber was evacuated to ≈10 mbar and hermetically sealed. In addition, a vacuum ($p<10^{-5}$ mbar) was maintained in an outer cell. The cooling and heating of the sample was accomplished with a constant flow of liquid nitrogen and with an electric resistor. With this setup a temperature stability better than 0.02 K was achieved.

Results: We have shown that capillary waves freeze on glycerol surfaces when the material is transformed to the supercooled state [4]. This freezing occurs at much higher temperatures than the calorimetric glass transition temperature T_g of the bulk material.

Conclusions: We have found that our results do not mean that T_g is altered at the surface since capillary waves are quasi long-range fluctuations in sharp contrast to the short ranged fluctuations in a liquid, which extend on length scales on the order of the bulk correlation length.

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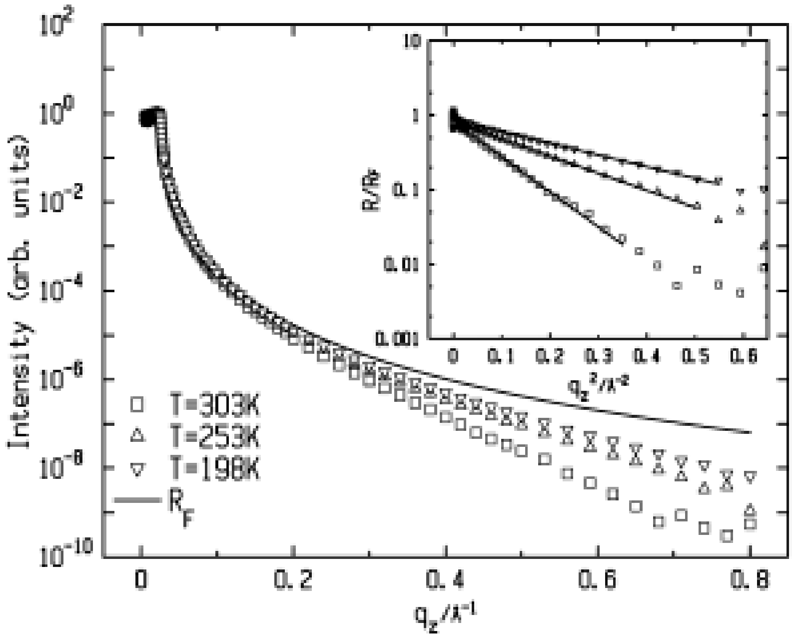

Fig. 1 Examples of reflectivity scans on a glycerol surface at different temperatures. The background arising from bulk scattering has been subtracted. The solid line is the Fresnel reflectivity R_F of an ideally smooth surface. The inset shows the data normalized to R_F. The slopes of the lines fitted to the data in the inset are proportional to the square of the corresponding rms surface roughness.